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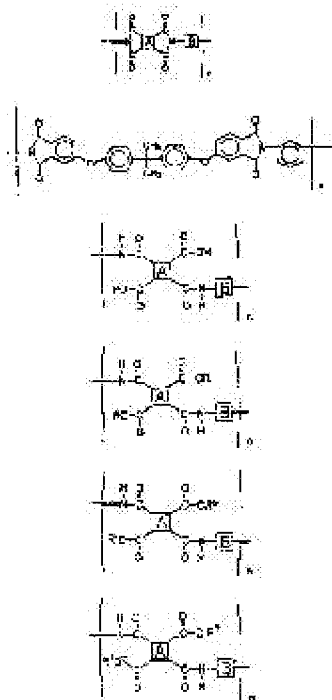
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(54) **ELECTROLUMINESCENT ELEMENT HAVING ORGANIC THIN FILM
LAYER CONTAINING POLYIMIDE**



(57)Abstract:

PROBLEM TO BE SOLVED: To provide an electric light emitting element which excels in heat resistance and mechanical property, improves the stability, and ensures long service life by constituting a transparent electrode layer, a metal electrode layer, and an organic intermediate layer positioned between two electrode layers by adhering closely thereto by an organic light emitting layer, a selective hole transfer layer, and a selective electron transfer layer and making them contain specific polyimide.

SOLUTION: The polyimide used has a structure expressed in formula I, and A denotes a residue derived from a dianhydride, B denotes a residue induced from diamine compound, and (n) denotes an integer of 2 or more. Moreover, it is preferable that polyetherimide or polyimide in formula II is used by heating a poly(amic acid) in formula III, a polyamic acid dialkyl ester in formula IV, a

covalent photosensitive polyimide precursor in formula V, or an ionic bond polyimide precursor in formula VI, and turning them into imide. A, B and (n) represent the same as defined in formula I, R denotes an alkyl group, R* denotes a covalent photosensitive functional group, and R+ denotes an ionic bond photosensitive functional group.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to an organic electrophotoluminescence element. More particularly, an organic layer is related with the organic electrophotoluminescence element which has a polyimide layer with luminescence and/or mediated transport capability, and has the improved luminous efficiency, the outstanding stability, and a long life.

[0002]

[Description of the Prior Art]Generally, an electrophotoluminescence element has a laminated structure which consists of an organic layer containing the organic luminous

layer located between a transparent electrode, a metal electrode, and said two electrodes, and drives it by exchange or direct current. In a direct-current drive, in a transparent electrode, the anode and a metal electrode act on the negative pole. In order to raise electrophotoluminescence efficiency, an organic layer may contain further an electron hole transporting bed and an electron transport layer in the form of multilayer structure. [0003]For example, in a direct-current drive, it is stuck to a special electron hole transporting bed between the anode and an organic luminous layer, and it may be located. An alternative electron transport layer can be inserted between catholyte and an organic luminous layer. Therefore, with the organic substance used, the organic layer of an electrophotoluminescence element may be a monolayer, two-layer, or three layers, and each layer contains the various organic photogene, electron hole transportation substances, and electron transport substances of combination. As the anode, an indium-tin-oxide glass plate is mainly used, and the metal layer of magnesium, aluminum, indium, or silver-magnesium is used as the negative pole.

[0004]However, when vibration, crystallization, and spreading diffusion arose for the Joule heat generated when driving an element, the organic layer which manufactured the single molecule in accordance with the usual vacuum deposition method, for example, an electron hole transfer layer, had a problem to which a thin film is changed or destroyed and the life of an element becomes short. The polymethylmethacrylate (PMMA) whose glass transition temperature is 105 °C these days in order to solve such a problem, Polycarbonate (PC) or glass transition temperature whose glass transition temperature is 145 °C made the matrix of other vinyl system polymers 150 °C or less distribute an active substance, for example, an electron hole transmitter, and organic photogene, and manufactured the organic layer. however, The stability of the organic layer manufactured since these polymers had low heat resistance is a satisfying thing. As for nothing, there is °C (Kido et al., Appl. Phys. Lett., 61, No. 7, 171, 1992; and Jpn, J.Appl. Phys., 31, No.78, L960, 1992 references).

[0005]On the other hand, U.S. Pat. No. 5,609,970, No. 5,571,626, No. 5,414,069, and electrophotoluminescence polymers with new No. 5,376,456 are indicated. However, composition of such special polymers takes many expenses, and since the synthetic course is complicated, mass production of an electrophotoluminescence element is not suited.

[0006]The polyimide system compound has been widely used in the electrical and electric equipment and the electronic field. For example, using as package INGU of an electrophotoluminescence element or an insulation material is reported (refer to U.S. Pat. No. 5,505,985 and No. 5,416,622). Using polyimide as an electron hole transporting bed is reported to Japan JP,4-93389,A.

It is indicated by JP,7-230881,A if polyimide containing the silicon heat-treated and manufactured at the temperature of 500 thru/or 1,000 °C has electron hole transport capacity.

However, the driver voltage of the last element is still high.

[0007]Therefore, stability and electrophotoluminescence efficiency are improved and development of an electrophotoluminescence element with sufficient economical efficiency is required.

[0008]

[Problem(s) to be Solved by the Invention]Therefore, the purpose of this invention is to

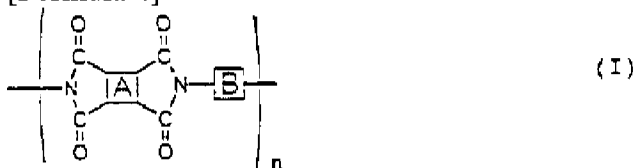
provide the electrophotoluminescence element which it excels in heat resistance and mechanical properties, and stability is improved, and has a long life.

[0009]

[Means for Solving the Problem]In an electrophotoluminescence element including an organic intermediate layer of this invention therefore stuck and located like 1 operative condition between a transparent electrode layer, a metal electrode layer, and said two electrode layers, Said organic intermediate layer consists of an organic luminous layer, an alternative electron hole transporting bed, and an alternative electron transport layer, and an electrophotoluminescence element containing polyimide of following general formula (I) is provided.

[0010]

[Formula 4]



The residue to which A was derived from dianhydride by said formula, the residue to which B was derived from the diamine compound, and n are two or more integers.

[0011]The organic luminous layer which contains a transparent positive electrode layer, a metal cathode layer, and organic photogene according to other embodiments of this invention, Said organic luminous layer is stuck and located between the whole surface of catholyte and an electron hole transporting bed including an electron hole transporting bed, and the electrophotoluminescence element for which the other sides of said electron hole transporting bed contain the electron hole transportation substance in which said electron hole transporting bed was distributed by the polyimide matrix in the anode and the electrophotoluminescence element which has stuck is provided.

[0012]In the electrophotoluminescence element to which said organic luminous layer is stuck and located between catholyte and a positive electrode layer according to other embodiments of this invention again including a transparent positive electrode layer, a metal cathode layer, and the organic luminous layer containing organic photogene, The electrophotoluminescence element containing the organic photogene in which said organic luminous layer was distributed by the polyimide matrix is provided.

[0013]

[Embodiment of the Invention]Hereafter, this invention is explained still in detail.

[0014]This invention is characterized by the electrophotoluminescence element by one mode comprising the following.

The anode which etched and manufactured the indium-tin-oxide (below indium tin oxide; calls it ITO) layer coated on a transparent substrate like glass by the usual method according to the pattern determined beforehand.

The metal electrode layer manufactured with aluminum, magnesium, calcium, silver, other metal, or an alloy.

The organic intermediate layer containing the organic luminous layer stuck and located between said transparent electrode layer and a metal electrode layer.

The electrophotoluminescence element of this invention can operate with exchange or a direct current. In a direct-current operation, a transparent electrode turns into the anode

and a metal electrode acts as the negative pole.

[0015] Said organic intermediate layer may contain further the electron hole transporting bed inserted between the transparent electrode layer and the organic luminous layer. An organic intermediate layer may contain further the electron transport layer inserted between the metal electrode layer and the organic luminous layer.

[0016] Therefore, the organic intermediate layer can manufacture with the gestalt of a monolayer or multilayer structure by in what kind of combination organic photogene, an electron hole transportation substance, and an electron transport substance are used. For example, an organic intermediate layer may be a bilayer gestalt which consists of electron hole transportation / organic luminous layer and an electron transport layer or an electron hole transporting bed, and organic luminescence / electron transport layer. An organic intermediate layer may also be a three-layer gestalt which consists of an electron hole transporting bed, an organic luminous layer, and an electron transport layer.

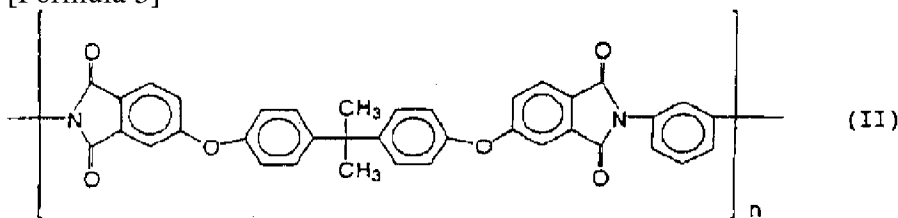
[0017] The feature of this invention is using polyimide for an organic intermediate layer.

[0018] Polyimide of general formula (I) shows electron hole transportation, luminescence, or electron transportation capability, when either of the residue of A and B contains a functional group with electron hole transportation, luminescence, or electron transportation capability.

[0019] Insoluble polyimide and glass transition temperature which heat-treated the polyimide precursor and were derived are not less than 220 **, and the illustration polyimide which may be used for this invention is soluble polyimide [like the polyether imide of following general formula (II) which is 40,000 or more] whose molecular weight is.

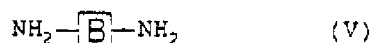
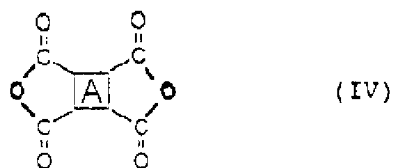
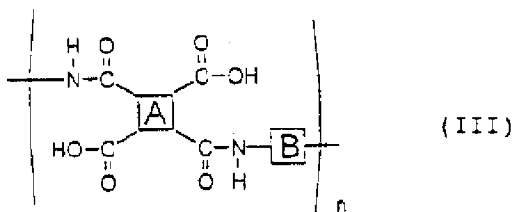
[0020]

[Formula 5]



By said formula, n is two or more integers.

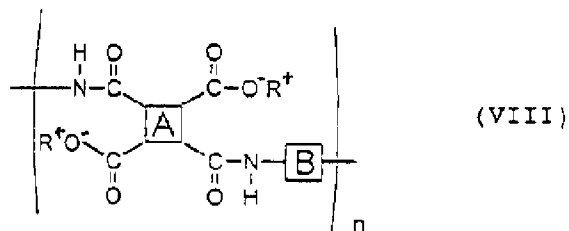
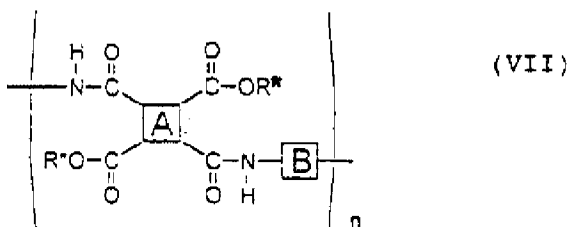
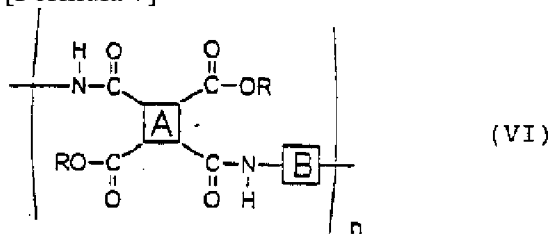
[0021] The polyimide precursor which may be used for this invention N-methyl-2-pyrrolidone, Dimethylacetamide, a tetrahydrofuran, dimethylformamide, They are 24 thru/or the poly AMIKU acid (polyamic acid) of general formula (III) produced by carrying out 48 time processings at the temperature of 0 thru/or 60 ** about dianhydride of general formula (IV), and the mixture of diamine of general formula (V) in solvents, such as dimethyl sulfoxide (DMSO). : [Formula 6]



Poly AMIKU acid dialkyl ester of general formula (VI), covalent bond type photosensitive polyimide precursors of general formula (VII), and ionic bond type photosensitive polyimide precursors of general formula (VIII) can also be used. A polyimide precursor may be converted into polyimide which ***** in general formula (I) by forming heat imide at temperature of the 100 thru/or 600 ** range, or irradiating with ultraviolet rays.

[0022]

[Formula 7]



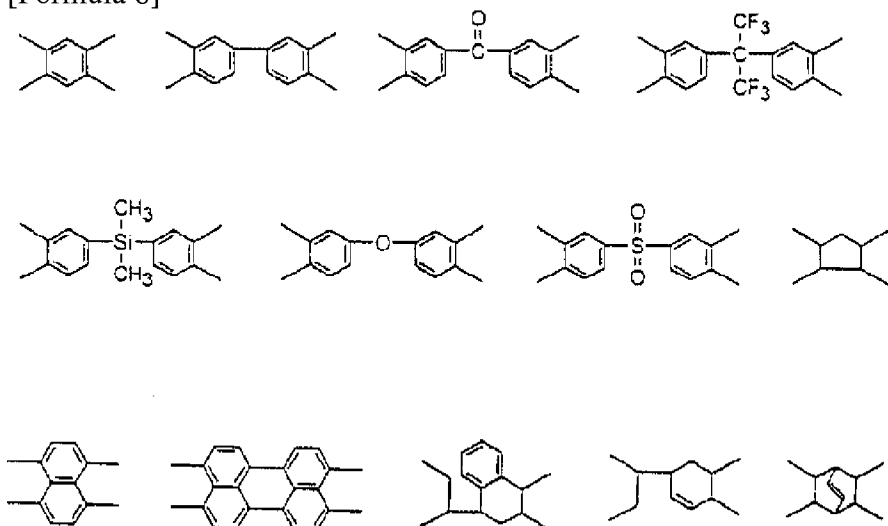
By said formula, A, B, and n are as [said] having given the definition, and R An alkyl

group, R* A covalent bond type photosensitivity functional group like $-\text{CH}_2\text{CH}_2\text{OC}(=\text{O})\text{CH}=\text{CH}_2$, R⁺ is an ionic bond type photosensitivity functional group like $^+\text{NH}(\text{CH}_3)_2\text{CH}_2\text{CH}_2\text{OC}(=\text{O})\text{CH}=\text{CH}-\text{Ph}-\text{N}_3$, and n is 30 thru/or 150.

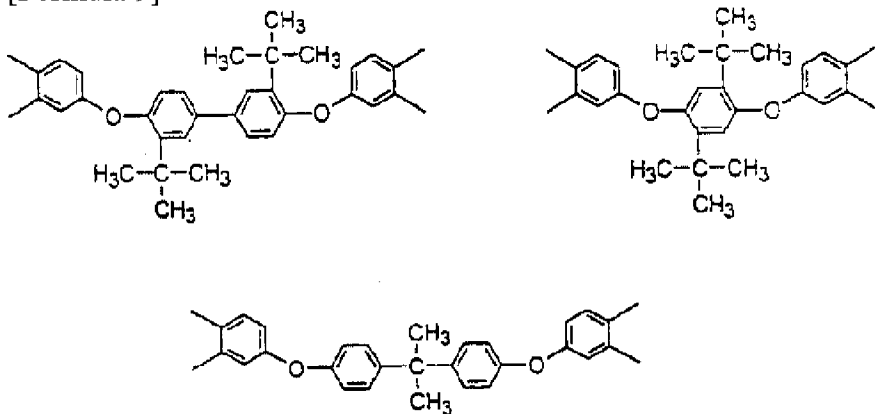
[0023] To dianhydride of general formula (IV) used for deriving A residue by said formula. Pyromellitic dianhydride, 3,4,3',4'-biphenyl tetracarboxylic dianhydride, 3,3',4,4'-benzophenone tetracarboxylic dianhydride, 4,4'-(hexafluoro propylidene) JI phthalic anhydride, 4,4'-(dimethylsilicon) JI phthalic anhydride, 4,4'-oxydi phthalic anhydride, 3,3',4,4'-diphenylsulfone tetracarboxylic dianhydride, 1,2,3,4-cyclopentane tetracarboxylic dianhydride, the naphthalene- 1, 4, and 5, 8-tetracarboxylic dianhydride, 3,4,9,10-perylene tetracarboxylic dianhydride, the 4-(2,5-dioxotetrahydrofuran 3-yl) tetralin 1, 2-dicarboxylic anhydride, The 5-(2,5-dioxotetrahydrofuryl)-3-methyl-3-cyclohexene 1, 2-dicarboxylic anhydride, Bicyclo[2,2,2] oct-7-ene- 2,3,5,6-tetracarboxylic dianhydride, a 2,2'-di-t-butylbiphenyl screw (phthalic anhydride ether), a 2,5'-di-t-butylphenyl screw (phthalic anhydride ether), And a bisphenol A-screw (phthalic anhydride ether) etc. are contained. The structure of A of polyimide of general formula (I) derived from said dianhydride is as follows.

[0024]

[Formula 8]



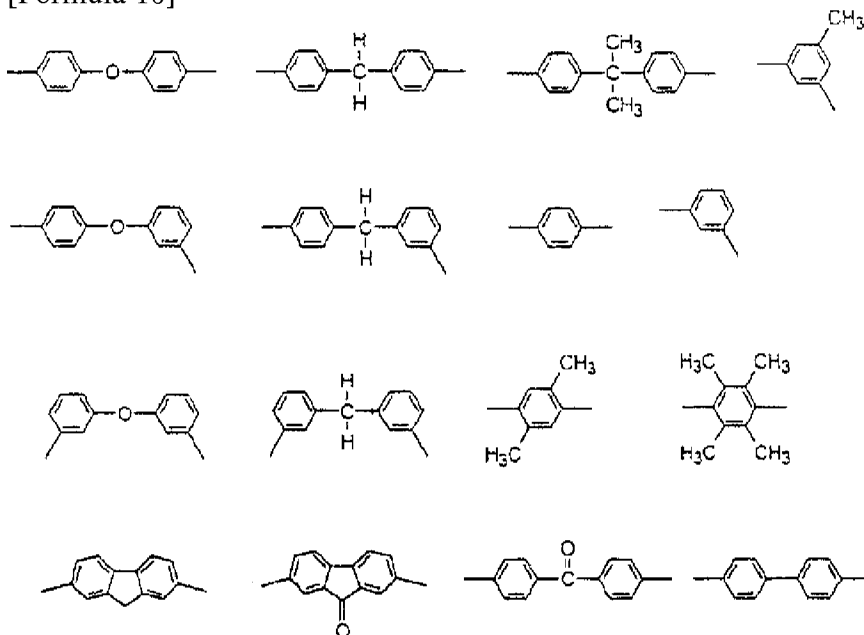
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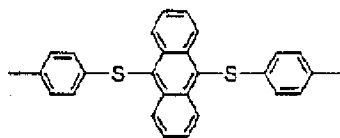
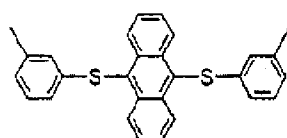
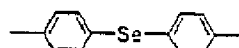
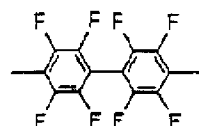
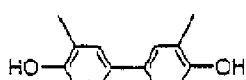
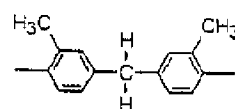
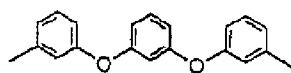
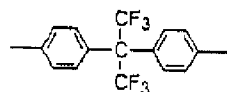
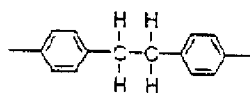
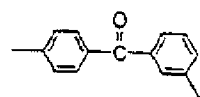
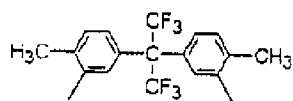
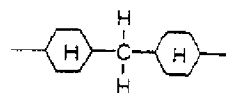
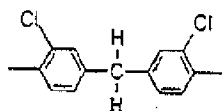
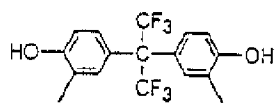
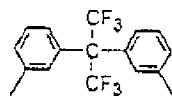
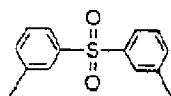
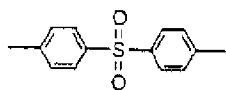
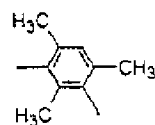
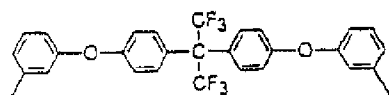
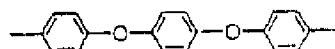
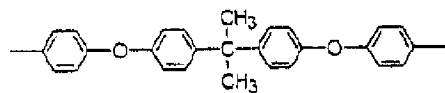
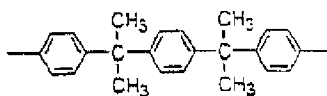
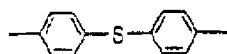
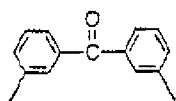
As an example of diamine of general formula (V) which forms the residue B, 4,4'-diaminodiphenyl ether, 4,4'-diaminodiphenylmethane, 2,2-bis(4-aminophenyl)propane, 3,5-diaminotoluene, 3,4-diaminodiphenyl ether, 3,4'-diaminodiphenylmethane, A 4,4'-phenylenediamine, a 3,4-phenylenediamine, 3,3'-diaminodiphenyl ether, 3,3'-diaminodiphenylmethane, 2,5-dimethyl- p-phenylene diamine, 2,3,5,6-tetramethyl p-phenylene diamine, a diaminofluorene, Diamino fluorenone, 4,4'-diaminobenzophenone, 4,4'-diaminobiphenyl, 3,3'-diaminobenzophenone, a 4,4'-diaminosulfide, alpha and alpha'-bis(4-aminophenyl)-1,4-diisopropylbenzene, 2,2-bis[4-(4-aminophenoxy) phenyl] propane, 1,4-bis(4-aminophenoxy)benzene, 2,2-bis[4-(3-aminophenoxy) phenyl] hexafluoropropane, 2,6-diaminotoluene, mesitylenediamine, 4,4'-diaminodiphenyl sulfone, 3,3'-diaminodiphenyl sulfone, 3,3'-bis(aminophenyl)hexafluoropropane, 2,2-bis(3-amino-4-hydroxyphenyl)hexafluoropropane, 4,4'-bis(2-chloroanilino)methane, 4,4-bis(aminocyclohexyl)methane, 2,2'-bis(3-amino-4-methylphenyl)hexafluoropropane, 3,4'-diaminobenzophenone, 4,4'-diaminobibenzyl, 4,4'-bis(aminophenyl)hexafluoropropane, 1,3'-bis(m-aminophenoxy)benzene, 4,4'-methylene-bis-o-toluidine, 3,3'-diamino-4,4'-dihydroxybiphenyl, 4,4'-diaminooctafluoro diphenyl, a 4,4'-bis(aminophenyl)selenide, 9,10-bis(3-amino phenylthio)anthracene, 9,10-bis(4-amino phenylthio)anthracene, 9,10-bis(3-aminoanilino)anthracene, 9,10-bis(4-aminoanilino)anthracene, the acridine yellow G, acriflavine, a 3,6-diaminoacridine, A 6,9-diamino-2-ethoxyacridine, the basic fuchsin, the methylation basic fuchsin, 2,4-diamino-6-phenyl-1,3,5-triazine, 9,10-diaminophenanthrene, There are 3,8-diamino-6-phenyl phenanthridine, dimidium bromide, an ethidium bromide, iodination propidium, thionine, 3,7-diamino-5-phenyl FENAJIUMU chloride, 3,3'-dimethylnaphthidine, etc. The structure of B of polyimide of general formula (I) derived from said diamine is as follows.

[0025]

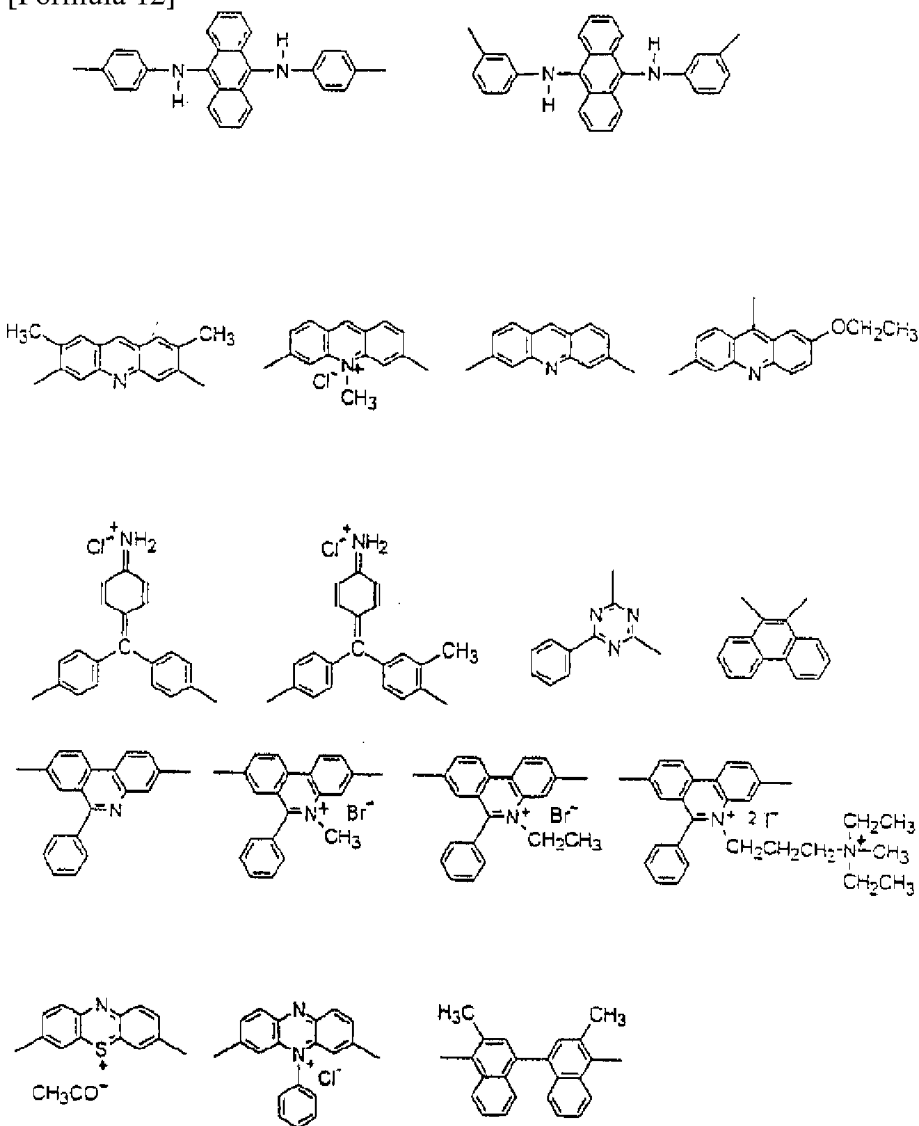
[Formula 10]



[Formula 11]



[Formula 12]



The electron hole transporting bed of electron hole transporting bed this invention contains the mixture of polyimide of general formula (I) which has residue with electron hole communicative competence or polyimide, and a usual electron hole transportation substance like the quality of a nitrogen inclusion. As an example of a concrete transportation substance, there are N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD) and poly (N-vinylcarbazole), among these TPD is preferred. [0026]a case where an electron hole transportation substance is distributed by polyether imide (below polyetherimide: calls it PEI) of general formula (II) -- a weight ratio of a polyether imide:electron hole transportation substance -- 90:10 thru/or 10:90 -- it is 50:50 preferably. If a weight ratio is less than 90:10, and electron hole transport capacity will become low and will exceed 90:10, the surface homogeneity of an electron hole transporting bed will fall. A solvent is made to distribute a suitable mixture of PEI and an electron hole transportation substance by 0.3 thru/or 10% of the weight of concentration, and a coating solution is manufactured. Under the present circumstances, a solvent can be

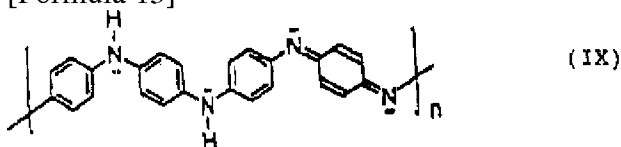
chosen from chloroform, dichloromethane, dimethylformamide, dimethylacetamide, N-methyl-2-pyrrolidone (NMP), tetrahydrofurans, and these mixtures.

[0027] A coating solution of manufactured PEI / electron hole transportation substance is coated on a transparent electrode by a usual wet process like spin coating, dock TABU lading, and screen printing. For example, said solution is coated with 500 thru/or 8,000 rpm for 1 thru/or 5 minutes on the ITO anode, at 30 thru/or 100 **, 1 minute cannot be found and a coating layer is dried for 1 hour.

[0028] A polymers electron hole transportation substance may be used by one mode of this invention. For example, doped poly aniline which doped and manufactured a poly aniline EMERARU gin base (PANI-EB) of following structural-formula (IX) from acid is blended with polyimide, and an electron hole transporting bed can be manufactured.

[0029]

[Formula 13]

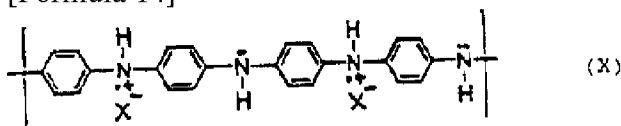


By said formula, n is two or more integers.

[0030] In chloroform, N-methyl-2-pyrrolidone (NMP), dimethylacetamide (DMAc), dimethylformamide (DMF), dimethyl sulfoxide (DMSO), and a solvent like a tetrahydrofuran (THF), A poly aniline EMERARU gin base Dodecylbenzenesulfonic acid (DBSA), It can dope from acid, such as camphor sulfonic acid (CSA) ((+) and (-) mold), benzenesulfonic acid, p-toluenesulfonic acid (PTSA), and 5-sulfosalicylic acid (SSA). The poly aniline doped from acid has a repeating unit of general formula (X).

[0031]

[Formula 14]



By said formula, X⁻ is the negative ion of acid.

[0032] The mixture which distributed by the weight ratio of 2:98 thru/or 95:5 to the polyimide precursor, and generated the doped poly aniline is dissolved in a solvent by 0.5 thru/or 10% of the weight of concentration. In this case, it is preferred to use N-methyl-2-pyrrolidone (NMP), dimethylacetamide (DMAc), and a weak basic solvent like dimethyl sulfoxide (DMSO).

[0033] Spin coating of the doped poly aniline/the polyimide precursor solution is carried out on an anode transparent electrode for 1 thru/or 5 minutes at 500 thru/or 8,000 rpm. Subsequently, after drying the coated thin film 30 minutes or more at 30 thru/or 100 **, heat imide is formed at 100 thru/or 600 **, or it irradiates with ultraviolet rays, and is made to convert into polyimide.

[0034] The doped poly aniline can also be distributed to polyether imide (PEI) by the weight ratio of 5:95 thru/or 80:20. Melt the mixture of poly aniline/polyether imide (PEI) in which the suitable weight ratio was doped by 0.5 thru/or 10% of the weight of concentration in a solvent like chloroform, and a coating solution is manufactured, This is

coated with 500 thru/or 8,000 rpm on the anode for 1 thru/or 5 minutes by a usual wet process like spin coating. Subsequently, a coating layer is dried 30 minutes or more at 30 thru/or 100 **.

[0035]A range of thickness of said electron hole transporting bed is 20 thru/or 100 nm.

[0036]An organic luminous layer of organic luminous layer this invention consists of a mixture of polyimide of general formula (I) which has organic luminescence residue or polyimide of general formula (I), and the usual organic photogene. Concrete organic photogene Tris(8-hydroxyquinolinolato) aluminum (Alq_3), 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-Piran (DCM), 1,4-JISUCHIRIRU benzene, anthracene, tetracene, pent RASEN, Coronene, perylene, pyrene, bis(8-quinolinolato)zinc (II), 9,10-diphenylanthracene, tris (4,4,4-trifluoro-1-(2-thienyl)-1,3-butane JIONO)-1,10-phenanthroline europium (III) ($\text{Eu}(\text{TTFA})$ 3P hen), Tris. (2,4-penta JIONO)-1,10-phenanthroline terbium (III) ($\text{Tb}(\text{ACAC})$ 3P hen) and tris (4,4,4-trifluoro-1-(2-thienyl)-1,3-butane JIONO)-1,10-phenanthroline dysprosium. (III) ($\text{Dy}(\text{TTFA})$ 3P hen etc. are included.

[0037]Organic photogene may be distributed by a weight ratio of 5:95 thru/or 70:30 ranges in soluble polyether imide of general formula (II). If a weight ratio is less than 5:95, and luminescence capability will become low and will exceed 70:30, the surface homogeneity of an electron hole transporting bed will fall. Chloroform, dichloromethane, and a solvent like chloroethane are made to distribute organic photogene / PEI mixture by 0.3 thru/or 10% of the weight of concentration, and a coating solution is manufactured.

[0038]The organic photogene / polyether imide coating solution manufactured in this way are coated on the surface of an electron hole transporting bed, when an electron hole transporting bed is used on the anode using a usual wet process like spin coating. For example, said solution is coated with 500 thru/or 8,000 rpm for 1 thru/or 5 minutes, and a generated coating layer is dried at 30 thru/or 100 ** for 30 thru/or 2 hours.

[0039]When a polyimide precursor is used for an organic luminous layer, Organic photogene and a polyimide precursor mixture of a weight ratio of 5:95 thru/or 70:30 are distributed to solvents, such as N-methyl-2-pyrrolidone (NMP), dimethylacetamide (DMAc), and dimethylformamide (DMF).

[0040]The organic photogene / polyimide precursor coating solution manufactured in this way are coated on the surface of an electron hole transporting bed, when an electron hole transporting bed is used on the anode using a usual wet process like spin coating. For example, after coating said solution with 500 thru/or 8,000 rpm 3 minutes or more and drying a generated coating layer at 40 thru/or 100 ** for 1 hour or more, heat amidation is carried out at 100 thru/or 600 ** for 1 thru/or 2 hours, or it irradiates with ultraviolet rays, and an organic luminous layer is manufactured.

[0041]As for thickness of said organic luminous layer, it is preferred that it is 10 thru/or 200 nm. Turn-on (turn-on) voltage of this invention can be adjusted by adjusting thickness of an organic luminous layer.

[0042]A mixture of polyimide of general formula (I) which has electron transportation residue or polyimide of general formula (I), and the usual electron transport substance may be used for an electron transport layer of electron transport layer this invention. As a concrete example of an electron transport substance, there are 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (butyl- PBD) and an oxadiazole derivative (OXD). The electron transport layer can coat and manufacture an electron transport substance layer

which vapor-deposited and formed an electron transport substance on an organic luminous layer, or was distributed in a polyimide matrix according to a procedure similar to the time of manufacture of an organic luminous layer.

[0043]As the multilayer-structure above-mentioned was carried out, an organic layer of this invention containing organic photogene, an electron hole transportation substance, and/or an electron transport substance may be a monolayer or a multilayer gestalt.

[0044]For example, an electron hole transportation substance and organic photogene can be mixed in one organic layer by coating a compound of polyether imide, organic photogene, and an electron hole transportation substance on the anode, and manufacturing electron hole transportation / organic luminous layer.

[0045]An organic metallic compound which has both organic luminescence and electron transportation capability. For example, a polyimide matrix is made to distribute tris(8-hydroxyquinolinolato) aluminum (Alq_3) according to a procedure similar to the time of manufacture of an organic luminous layer, and organic luminescence / electron transportation monolayer can be formed.

[0046]A constant rate of organic photogene can be mixed to an electron hole transporting bed so that an electron hole may be smoothly poured into an organic luminous layer from an electron hole transfer layer in the case of a drive of an organic light emitting element. When an electron transport layer is used for an electrophotoluminescence element of this invention, organic photogene is added also to an electron transport layer, and contact nature can be improved.

[0047]The desirable mode of desirable mode this invention is as follows.

[0048]Said organic intermediate layer An organic luminous layer and N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine, An electrophotoluminescence element which consists of an electron hole transporting bed which contains polyether imide of an electron hole transportation substance chosen from a group which consists of poly (N-vinylcarbazole) and these mixtures, and general formula (II) by a weight ratio of 10:90 thru/or 90:10 ranges.

[0049]An electrophotoluminescence element for which said organic luminous layer contains organic photogene and said polyether imide by a weight ratio of 5:95 thru/or 70:30 ranges.

[0050]An electrophotoluminescence element which said organic intermediate layer becomes from an organic luminous layer and an electron hole transporting bed which contains three ingredients, said polyether imide, an electron hole transportation substance, and organic photogene, by a weight ratio of 4:3:3 thru/or 5:4:1 ranges.

[0051]An electrophotoluminescence element which consists of an electron hole transporting bed manufactured by said organic intermediate layer's coating a mixture of poly aniline doped from an organic luminous layer and acid of 2:98 thru/or a 95:5 weight-ratio range, and said polyimide precursor, and heating-imide-izing a coating layer.

[0052]An electrophotoluminescence element which said organic intermediate layer becomes from an organic luminous layer and an electron hole transporting bed which contains poly aniline doped from acid, and said polyether imide by a weight ratio of 5:95 thru/or 80:20 ranges.

[0053]An electrophotoluminescence element manufactured by said organic luminous layer's coating organic photogene of 2:98 thru/or a 95:5 weight-ratio range, and a mixture of said polyimide precursor, and heating-imide-izing a coating layer.

[0054]An electron hole transporting bed in which said organic intermediate layer contains an electron hole transportation substance and said polyether imide by a weight ratio of 5:95 thru/or 80:20 ranges, An electrophotoluminescence element which consists of an organic luminous layer which contains an organic metallic compound which has both organic luminescence and electron transportation capability, and said polyether imide by a weight ratio of 5:95 thru/or 70:30 ranges.

[0055]An electron hole transporting bed manufactured by said organic intermediate layer's coating a mixture of poly aniline by which 2:98 thru/or a 95:5 weight-ratio range were doped, and said polyimide precursor, and heating-imide-izing a coating layer, An electrophotoluminescence element which consists of an organic luminous layer which contains organic photogene and said polyether imide by a weight ratio of 5:95 thru/or 70:30 ranges.

[0056]In order to improve adhesion with metal and an organic layer at the same time it reduces contact resistance, on the anode or before coating the negative pole, a polyimide film can be further coated with this invention. Stability may be made to increase by carrying out packaging of the element of this invention by suitable resin, and protecting from air and moisture.

[0057]In an electrophotoluminescence element of this invention which has a monolayer and an organic intermediate layer of multilayer structure, since stability, electrophotoluminescence efficiency, and a life of an element are increased, polyimide may be used for one or more organic layers. In particular, when an organic intermediate layer is a gestalt of the two-layer structure of polyimide/polyimide, the physical stability of an element is improved dramatically. Quantity and impressed electromotive force of organic photogene which are distributed by polyimide are adjusted, and wavelength of luminescence can be adjusted.

[0058]Since an organic electrophotoluminescence element of this invention can be manufactured at a wet process, it can be manufactured in a monotonous large form. When one or more polyimide layers are used for all the organic layers and a flexible electrode is used, manufacture of a bent display device is also possible. This invention may be utilized not only for an organic electrophotoluminescence element but for a solar cell (solar cell), FET (field effect transistor), a photo diode, etc.

[0059]

[Example]Hereafter, the following example explains this invention still in detail. However, it is only illustrating this invention and the following example does not restrict the range of this invention.

[0060]ITO was coated on example 1 clear glass, and the transparent positive electrode was manufactured.

[0061]N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (below N, N'-diphenyl-N, and N'-bis(3-methylphenyl)-4, 4'-diamine:.) And it called it TPD, it dissolved in chloroform by 0.5% of the weight of concentration by making polyether imide (PEI) of general formula (II) into the weight ratio 50:50. Said transparent ITO anode was coated with said TPD/PEI solution, it dried and the electron hole transporting bed of 60-nm thickness was manufactured.

[0062]Vacuum deposition of the tris(8-hydroxyquinolinolato) aluminum (below Tris(8-hydroxyquinolinolato) aluminium: calls it Alq₃) was carried out on said electron hole transporting bed, and organic luminescence / electron transport layer of 10-nm thickness

were manufactured.

[0063]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of a 2×10^{-6} torr on said organic luminescence / electron transport layer, catholyte was manufactured, on said negative pole, indium was vapor-deposited and the conductive protective layer was manufactured.

[0064]As for 520 nm and the turn-on (turn-on) voltage of the element, 9V and operating potential of the luminous wavelength were 9-15V.

[0065]ITO was coated on example 2 clear glass, and the transparent positive electrode was manufactured.

[0066]Polyether imide (PEI) of N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD) and general formula (II) is made into the weight ratio 50:50. It stirred in chloroform for 24 hours, and the TPD/PEI solution of 1.0% of the weight of concentration was obtained. Spin coating of said TPD/PEI solution was carried out for 3 minutes at 4,000 rpm on said transparent ITO anode. The coated thin film was dried at the temperature of the 45 thru/or 50 °C range, and the electron hole transporting bed of 40-nm thickness was manufactured.

[0067]The polyether imide (PEI) of tris(8-hydroxyquinolinolato) aluminum (Alq_3) and general formula (II) was mixed by the weight ratio of 50:50, the mixture was stirred in chloroform for 24 hours, and the Alq_3 /PEI solution of 0.5% of the weight of concentration was obtained. Spin coating of said solution was carried out for 3 minutes at 3,000 rpm on said transparent ITO anode. The coated thin film was dried at the temperature of the 50 °C range for 1 hour, and organic luminescence / electron transport layer of 25-nm thickness were manufactured.

[0068]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of 10^{-6} torr on said organic luminescence / electron transport layer, and 400-nm catholyte was manufactured.

[0069]As for 520 nm and the turn-on voltage of the element, 9V and operating potential of the luminous wavelength were 9-14V.

[0070]ITO was coated on example 3 clear glass, and the transparent positive electrode was manufactured.

[0071]The polyether imide (PEI) (molecular weight 45,000) of general formula (II), N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD) and tris(8-hydroxyquinolinolato) aluminum (Alq_3) were mixed by the weight ratio of 50:40:10. The generated mixture was stirred in chloroform for 24 hours, and the TPD+ Alq_3 /PEI solution of the concentration of 0.5% weight was obtained. Spin coating of said TPD+ Alq_3 /PEI solution was carried out for 1 minute at 5,000 rpm on said transparent ITO anode. The coated thin film was dried at 60 °C for 1 hour, and the electron hole transporting bed of 30-nm thickness was manufactured.

[0072]Vacuum deposition of the tris(8-hydroxyquinolinolato) aluminum (Alq_3) was carried out on said electron hole transporting bed, and organic luminescence / electron transport layer was manufactured.

[0073]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of 10^{-6} torr on said organic luminescence / electron transport layer, and 400-nm catholyte was manufactured.

[0074]As for 520 nm and the turn-on voltage of the element, 9V and operating potential of the luminous wavelength were 9-12V.

[0075]ITO was coated on example 4 clear glass, and the transparent positive electrode was manufactured.

[0076]Poly aniline EMERARU gin base (below Polyaniline emeraldine base:.) PANI -- saying -- the poly aniline (PANI-DBSA) doped by doping with the dodecylbenzenesulfonic acid (below Dodecylbenzene sulfonic acid: calls it DBSA) which has melted into N-methyl-2-pyrrolidone was obtained. The doped poly aniline (PANI-DBSA) was mixed by the polyether imide (PEI) of general formula (II), and the weight ratio of 50:50, it stirred in chloroform for 24 hours, and the PANI-DBSA/PEI solution of 0.5% of the weight of concentration was obtained. Spin coating of the PANI-DBSA/PEI solution was carried out for 90 seconds at 5,000 rpm on said transparent ITO anode. The coated thin film was dried at 60 °C for 1 hour, and the electron hole transporting bed of 50-nm thickness was manufactured.

[0077]5 Vacuum deposition of the tris(8-hydroxyquinolinolato) aluminum (Alq_3) was carried out on said electron hole transporting bed under the pressure of 5×10^{-6} torr, and organic luminescence / electron transport layer of 10-nm thickness were manufactured.

[0078]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of 5×10^{-6} torr (torr) on said organic luminescence / electron transport layer, and catholyte was manufactured.

[0079]As for 520 nm and the turn-on voltage of the element, 6V and operating potential of the luminous wavelength were 6-9V.

[0080]ITO was coated on example 5 clear glass, and the transparent positive electrode was manufactured.

[0081]The poly aniline (PANI-DBSA) doped by doping a poly aniline EMERARU gin base (PANI) with the dodecylbenzenesulfonic acid (DBSA) which has melted into N-methyl-2-pyrrolidone was obtained.

[0082]A 4,4-oxo phenylenediamine and pyromellitic dianhydride were made to react at 25 °C for 48 hours, and the polyimide precursor, i.e., poly AMIKU acid of general formula (III), was manufactured.

[0083]Poly aniline (PANI-DBSA) and poly AMIKU acid (below Polyamic acid: calls it PAA) which were doped were mixed by the weight ratio of 20:80, it stirred in chloroform, and the PANI-DBSA/PAA solution of concentration was obtained 1% of the weight. Spin coating of said PANI-DBSA/PAA solution was carried out for 3 minutes at 5,000 rpm on said transparent ITO anode. The coated thin film was formed into heat imide at 200 °C for 1 hour, and the electron hole transporting bed of 60-nm thickness was manufactured.

[0084]On said electron hole transporting bed, tris(8-hydroxyquinolinolato) aluminum (Alq_3) was vapor-deposited at 0.02nm/second in speed under the pressure of 5×10^{-6} torr, and organic luminescence / electron transport layer of 5-nm thickness were manufactured.

[0085]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of 5×10^{-6} torr (torr) on said organic luminescence / electron transport layer, and 300-nm catholyte was manufactured.

[0086]As for 520 nm and the turn-on voltage of the element, 4V and operating potential of the luminous wavelength were 4-8V.

[0087]ITO was coated on example 6 clear glass, and the transparent positive electrode was manufactured.

[0088]Poly AMIKU acid (PAA) of general formula (III) manufactured in N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD) and Example 5 was mixed by the weight ratio of 30:70. The mixture was melted in dimethylformamide and the TPD/PAA solution of 1% of the weight of concentration was obtained. Spin coating of said solution was carried out for 3 minutes at 3,000 rpm on said transparent ITO anode. Heating amidation of the coated thin film was carried out at 200 °C for 1 hour, and the electron hole transporting bed of 40-nm thickness was manufactured.

[0089]The polyether imide (PEI) of perylene and general formula (II) was mixed by the weight ratio of 30:70, the mixture was stirred in chloroform for 24 hours, and the perylene / PEI solution of 1% of the weight of concentration were obtained. The thin film which carried out spin coating for 3 minutes, and coated said solution with 3,000 rpm on said transparent ITO anode was dried at 47 °C for 1 hour, and the organic luminous layer of 35-nm thickness was manufactured.

[0090]Subsequently, vacuum deposition of the aluminum was carried out under the pressure of 10^{-6} torr (torr) on said organic luminescence, and 300-nm catholyte was manufactured.

[0091]As for 520 nm/610 nm, and the turn-on voltage of the element, 9V and operating potential of the luminous wavelength were 15-19V.

[0092]ITO was coated on example 7 clear glass, and the transparent positive electrode was manufactured.

[0093]The polyether imide (PEI) of 1,1,4,4-tetraphenyl-1,3-butadiene (below 1, 1, 4, and 4-tetraphenyl-1,3-butadiene: calls it TB) and general formula (II) was mixed by the weight ratio of 50:50. The mixture was melted by 2.0% of the weight of concentration in chloroform, and 2.0% of the weight of the TB/PEI solution was obtained. Spin coating of said solution was carried out for 3 minutes at 3,000 rpm at said transparent ITO anode. The coated thin film was dried at 50 °C for 1 hour, and the organic luminous layer of 100-nm thickness was manufactured.

[0094]Subsequently, aluminum was vapor-deposited under the pressure of 10^{-6} torr (torr) on the organic luminous layer, catholyte was manufactured, on said negative pole, indium was vapor-deposited and the conduction protective layer was manufactured.

[0095]As for 440 nm and the turn-on voltage of the element, 12V and operating potential of the luminous wavelength were 13V.

[0096]ITO was coated on example 8 clear glass, and the transparent positive electrode was manufactured.

[0097]4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-Pyran (below 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4 H-pyran:.) And it called it DCM, the polyether imide (PEI) of general formula (II) was mixed by the weight ratio of 50:50. DCM/PEI was dissolved in chloroform and spin coating of the solution was carried out for 3 minutes at 3,000 rpm at said transparent ITO anode. The coated thin film was dried at 50 °C for 1 hour, and the organic luminous layer was manufactured.

[0098]Subsequently, on said organic luminous layer, aluminum was vapor-deposited under the pressure of a 5×10^{-6} torr (torr), and the catholyte of 300-nm thickness was manufactured.

[0099]As for 620 nm and the turn-on voltage of the element, 15V and operating potential of the luminous wavelength were 10-15V.

[0100]ITO was coated on example 9 clear glass, and the transparent positive electrode

was manufactured.

[0101]The polyether imide (PEI) of 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-Piran (DCM) and general formula (II) was mixed by the weight ratio of 50:50. DCM/PEI was dissolved in chloroform and spin coating of the solution was carried out for 3 minutes at 2,000 rpm at said transparent ITO anode. The coated thin film was dried at 50 °C for 1 hour, and the organic luminous layer was manufactured.